Development of Theoretical Model for Effective Thermal Conductivity of Glass Microsphere Filled Polymer Composites

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Abstract

Thermal conductivity is an important thermal property of materials and plays an important role in determining their heat conduction/insulation capability. The present work aims to study the effect of embedded solid glass microspheres (SGM) on the effective thermal conductivity of epoxy resin. Composite samples were prepared by embedding SGMs of different sizes in the epoxy resin. Three-dimensional spheres-in-cube lattice array models were constructed using finite element method (FEM) to simulate the microstructure of composite materials for various SGM content ranging from 0 to about 17.9 vol %. Finally, guarded heat flow meter test method was used to measure the conductivity of these composites. A relevant theoretical model was deduced based on the law of minimal thermal resistance and the equal law of the specific thermal conductivity. The simulations were compared with k_{eff} values obtained from experiments and it is found that the FEM simulations and k_{eff} values of the theoretical model are fairly close to the measured k_{eff}. This study has shown that the embedment of glass microsphres results in substantial reduction of heat conductivity of epoxy resin and thereby improving its thermal insulation capability. Further, the size and content of SGMs would influence the extent of reduction of k_{eff}.

Keywords

Composites; Glass Microspheres; Thermal Conductivity; Insulation

Introduction

The improved performance of polymers and their composites in industrial and structural applications by the addition of solid filler materials has shown great promise and so it has lately been a subject of considerable interest. For many material applications, thermal properties of polymer composites serve as important parameter. An increase in temperature of approximately 10°C reduces the mean time to failure by a factor of two. Hence the thermal performance of moulded plastic packages is very important. Reinforced polymeric materials are being widely used in electronic systems due to their ease of manufacture, light weight and tailorable properties. Electronic systems produce a lot of heat during operation. As the size of packages becomes smaller, they encounter increasingly high temperatures. Variation in temperature adversely affects the reliability and electrical performance of the product. The application of composite materials in the electronic industry has renewed interest in the development of theoretical and experimental models for determination of the effective thermal conductivity of composite materials. Determination of the thermal conductivity of composite materials plays a crucial role in a number of industrial processes. Despite the importance of this material property and the considerable number of studies that have been carried out, the determination of effective thermal conductivity of a composite is partially understood. The effective thermal conductivity of a composite material is a complex function of their geometry, the thermal conductivity of the different phases, distribution within the medium and contact between the particles. Various kinds of polymers, and polymer matrix composites reinforced with metal particles have a wide range of industrial applications such as heaters, electrodes, composites with thermal durability at high temperature etc. These engineering composites are desired due to their low density, high corrosion resistance, ease of fabrication and low cost. Increasing use of polymer composites for various applications emphasizes its importance/significance in the thermal property analysis of an engineering system. Conductivity is one such important thermal property that needs to be evaluated for any new composite system. Generally, measuring the thermal conductivity accurately is helpful to study the heat transfer process and mechanisms in composite materials. Although it can be measured by experimental methods, analytical

methods and equations are often essential to predict thermal conductivities of composite materials. Considerable work has been reported on the subject of heat conductivity in polymers by Hansen and Ho, Peng and Landel, Choy and Young, Tavman etc. The fillers most frequently used are aluminum, copper and brass particles, short carbon fiber, carbon particles, graphite, aluminum nitride, magnetite particles etc. Progelhof et al. were the first to present an exhaustive overview on models and methods to predict the thermal conductivity of composite systems. Procter and Solc used Nielsen model as a prediction to investigate the thermal conductivity of several types of polymer composites filled with different fillers and confirmed its applicability. While Kumlutas and Tavman carried out a numerical and experimental study on thermal conductivity of particle filled polymer composites, Amar et al. reported the existence of a possible correlation between thermal conductivity and wear resistance of particulate filled composites. Recently, Nayak et.al. have reported on the modified thermal conductivity of pine wood dust filled epoxy-based composites.

The heat transfer process of porous materials is very complicated, especially for polymer composites. It is quite important, therefore, to understand the mechanisms of heat transfer in polymer composites which are potential insulating materials. For porous materials, several researchers have deduced effective thermal conductivity equations based on the Maxwell expression, or established a more accurate formula to calculate the effective thermal conductivity of such materials. The models proposed respectively by Nielsen, Agari and Uno, Dong et.al. and Cheng–Vachon can better estimate the effective thermal conductivity of filled composite materials, while the Agari– Nagai equation can predict for composites with high particle loading. While Liang and Qu analyzed the thermal conductivity of a porous material with closed spherical and cylindrical holes, Suvorov et al. studied the thermal conductivity of hollow emery filled composites. Foamed plastic is a polymeric material commonly used as thermal insulation but its application is limited considerably due to its poor mechanical properties. There is, therefore, a focus on the fabrication of a kind of reinforced polymeric system which is light but has better mechanical strength and good thermal insulation properties. In this context, rigid glass micro-spheres have some advantages as fillers in polymers such as low thermal conductivity, coefficient and density. In addition, these micro-particles do not generate stress concentration in the interface between the fillers and the matrix owing to their smooth spherical surface. These type of composites can be applied to building materials, space flight and aviation industry. Glass micro-spheres are preferred as fillers especially when composite properties such as isotropy or low melt viscosity are important. Moreover, the orientation effects associated with molding are minimal. There are only a few published papers on evaluation of effective thermal conductivity of polymer composites filled with glass beads Liang and Li reported on measurement of thermal conductivity of hollow glass-bead-filled polypropylene composites. Recently, they also made two-dimensional and three-dimensional finite element analysis on the heat transfer and simulated the variation of effective thermal conductivity of hollow glass microsphere filled polymer compositesand further studied the heat transfer in polymer composites filled with hollow glass micro-spheres and proposed a theoretical model to predict the thermal conductivity of such composite system. Later on Liang and Liu formulated a new theoretical model for the heat transfer in inorganic particulate-filled polymer composites. Yung et al reported on the preparation and properties of hollow glass microsphere-filled epoxy matrix composites. However, all these studies are for polymer composites filled with hollow glass spheres and surprisingly, and there is no report available on evaluation of effective thermal conductivity of solid glass microsphere filled polymer composites. In view of the above, the present work is undertaken to evaluate theoretically and experimentally the thermal conductivity of epoxy matrix composites filled solid glass micro-spheres. Epoxy is chosen primarily because it happens to be the most commonly used polymer and because of its low value of thermal conductivity (about 0.363 W/m-K). The objectives of this work includes the investigation of the heat transfer process in particulate-filled polymer composites to develop a new mathematical model for estimation of the effective thermal conductivity of composites and hence fabricating a new class of composites to further improve the insulating properties of epoxy by the incorporation of solid glass micro-spheres.

Many theoretical and empirical models have been proposed to predict the effective thermal conductivity of two-phase mixtures. Comprehensive review articles have discussed the applicability of many of these models. For a two-component composite, the simplest

alternatives would be with the materials arranged in either parallel or series with respect to heat flow, which gives the upper or lower bounds of effective thermal conductivity. For the parallel conduction model:

$$k_c = (1-\phi)k_m + \phi k_f \tag{1}$$

where k_c, k_m, k_f are the thermal conductivities of the composite, the matrix and the filler respectively and Φ_f is the volume fraction of filler.

For series conduction model:

$$\frac{1}{k_e} = \frac{1-\phi_f}{k_c} + \frac{\phi_f}{k_d} \tag{2}$$

The correlations presented by Eqs.(1) and (2) are derived on the basis of the rules-of-mixture. Tsao derived an equation related to the thermal conductivity of two-phase solid mixture to the conductivity of the individual components and to two parameters which describe the spatial distribution of the two phases. By assuming a parabolic distribution of the discontinuous phase in the continuous phase, Cheng and Vachon [5] obtained a solution to Tsao's [6] model that did not require knowledge of additional parameters. Agari and Uno [7] proposed a new model for filled polymers, which takes into account parallel and series conduction mechanisms. According to this model, the expression that governs the thermal conductivity of the composition is:

$$\log k_c = \phi \log c_1 k_f + (1-\phi)\log(c_2 k_m) \tag{3}$$

where C_1, C_2 are experimentally determined constants of order unity. C_1 is a measure of the effect of the particles on the secondary structure of the polymer, like crystallinity and the crystal size of the polymer. C_2 measures the ease of the particles to form conductive chains. Then, they modified the model to take into account the shape of the particles. Generally, this semi-empirical model seems to fit the experimental data well. However, adequate experimental data is needed for each type of composite in order to determine the necessary constants. For an infinitely dilute composite of spherical particles, the exact expression for the effective thermal conductivity is given by

$$\frac{k}{k_c} = 1 + 3\left\{\frac{k_d - k_c}{k_d + 2k_c}\right\} \tag{4}$$

where k, k_c and k_d are thermal conductivities of composite, continuous phase (matrix), and dispersed-phase (filler), respectively, and φ is the volume fraction of the dispersed-phase. Eq. is the well known Maxwell equation for dilute composites. The correlations presented by Eqs. 1 and 2 are derived on the basis of the Rules of Mixture (ROM). Lewis and Nielsen [Dilek Kumlutas,et.al. derived a semi-theoretical model by a modification of the Halpin-Tsai equation for a two-phase system:

$$k_c = k_m\left[\left(1 + AB\phi\right)/\left(1 - B\phi\psi\right)\right] \tag{5}$$

where,

$$\psi = 1 + \left\{(1-\phi_m)/\phi_m^2\right\}\phi$$

For an infinitely dilute composite of spherical filler particles, the exact expression for the effective thermal conductivity is given by Maxwell as:

$$\frac{k}{k_c} = 1 + 3\left(\frac{k_d - k_c}{k_d + 2k_c}\right) \tag{6}$$

where k, k_c and k_d are thermal conductivities of composite, continuous-phase (matrix), and dispersed-phase (filler), respectively and φ is the volume fraction of the dispersed-phase.

Russell obtained the conductivity using a series parallel network.

$$k_c = k_m\left[\phi^{2/3} + \frac{k_m}{k_f(1-\phi^{2/3})}\right] \Big/ \left[\phi^{2/3} - \phi + \frac{k_m}{k_f}(1+\phi-\phi^{2/3})\right] \tag{7}$$

An exhaustive review of the published literature reveals that most of the investigations are aimed at enhancing the thermal conductivity of the polymer rather than attempting to improve its insulation capabilities. Foamed plastic is a polymeric material commonly used for thermal insulation. However, its application is limited considerably due to its poor mechanical properties. There is, therefore, a focus on fabrication of a kind of reinforced polymeric system which is light but has better mechanical strength and good thermal insulation properties. Rigid glass micro-spheres have some advantages as fillers in polymers such as low thermal conductivity, coefficient and density. In addition, these micro-particles do not generate important stress concentration in the interface between the fillers and the matrix owing to their smooth spherical surface. These type of composites can be applied to building materials, space flight and aviation industry. Glass microspheres are preferred as fillers especially when composite properties such as isotropy or low melt viscosity are

important. Moreover, the orientation effects associated with molding are minimal.

Development of a Theoretical Model

H: side of the cube

r: radius of the spherical glass bead

k_p : thermal conductivities of polymer matrix

k_g : thermal conductivity of micro-sphere phase

Q: heat quantity

Q_p: heat quantity through the polymer matrix

Q_g: heat quantity through micro-sphere shell

h_1: H-2r

A: total area of cross-section (heat transfer area)

A_g: cross-sectional area of the micro-sphere

A_p: cross-sectional area of the polymer

V_p: volume of the polymer matrix

V_g: volume of the micro-sphere shell respectively

V_c: volume of the composite

ϱ_p: density of the polymer matrix phase

ϱ_g: density of microsphere phase

ϱ_c: density of the composite

R_1: heat resistance of Part I

R_2: heat resistance of Part II

R: total resistance

v_f: volume fraction of the filler in the matrix

T: temperature

The heat quantity through a body depends upon the route of heat transfer in the materials. In this article, a series model of heat transfer is considered. Fig.1 shows a series model of heat transfer in an inorganic particle-filled polymer composite. As our filler material is soild glass micro-spheres which are generally spherical in shape, therefore for convenience only the case of spherical inclusions is considered in the matrix. The element is divided into three parts, that is, part one, part two and part three with thermal conductivities k_1, k_2 and k_3, respectively. A_p and A_g are the cross-sectional areas of the resin matrix and filler, and Q_p and Q_g are the heat quantities through the areas of the matrix and filler individually. If it is supposed that a composite is made up of a number of squared elements, and each element contains only a spherical particle at its centre, the heat flows through the particle.

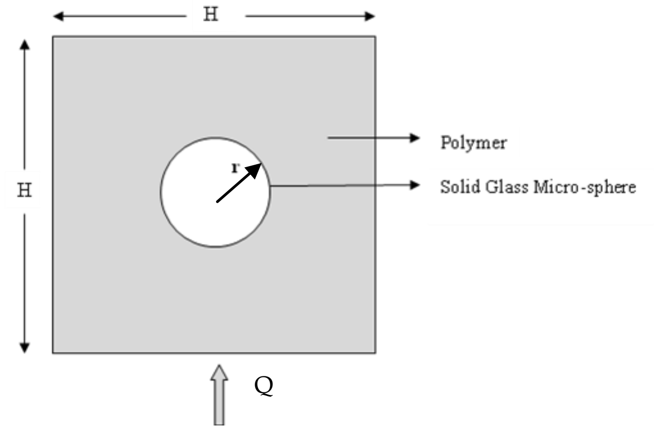


FIG.1 A HEAT TRANSFER MODEL IN PARTICULATE-FILLED POLYMER COMPOSITE

The theoritical analysis of the heat transfer in this paper is based on the following suppositions: (a) the distribution or dispersion of the solid micro-spheres in the polymer matrix is uniform and (b) the temperature distribution along the direction of heat flow is linear. An element from the composite is selected for analysis which is a straight cube with side length of H and there is a solid glass micro-sphere of radius (r) in the element.

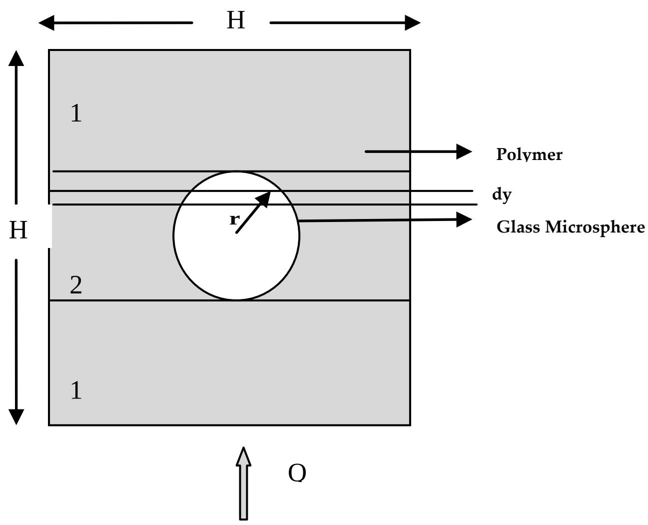


FIG.2 A SERIES MODEL OF HEAT TRANSFER IN PARTICULATE-FILLED COMPOSITE

The element is divided into polymer phase and micro-sphere phase. The heat quantity Q transfers from bottom to the top. The heat transport in solid glass micro-sphere filled polymer composites has two mechanisms: (i) solid thermal conduction and (ii) heat radiation on the surface between neighbouring particles. Polymer composite works usually under lower temperature conditions where the proportion of the thermal radiation in the total heat transfer is very small, hence the thermal radiation is neglected.

Part I:

$$k_1 = k_p \tag{8}$$

Part II: Taking a thin piece with thickness of dy (as shown in Fig.2), according to Fourier's theorem, k_2 is given by:

$$k_2 = \frac{Q_p+Q_g}{\left(\frac{dT}{dy}\right)S} = k_p\frac{Ap}{A} + k_g\frac{A_g}{A} \tag{9}$$

where, T is the temperature, A is the area of whole cross-section. k_p, k_g are the thermal conductivities of polymer matrix phase and micro-sphere phase. A_p and A_g are the cross-sectional areas of the polymer matrix and micro-sphere. Q_p and Q_g are the heat quantities through the polymer matrix and micro-spheres respectively. As the temperature distribution is linear, the average thermal conductivity of each section may first be obtained:

Part I:

$$k_1 = \int_{h_1} k_1\, dy/h_1) = k_p \tag{10}$$

Part II:

$$k_2 = (1/h_2)\int_{h_2} k_p \ (A_p/S)\ k_g(\ A_g/A)dy$$

$$= \left(\frac{1}{h_2A}\right)(k_pV_p + k_gV_g)$$

$$= \left(\frac{k_p V_p}{2Ar}\right) + \left(\frac{k_g V_g}{2Ar}\right) \tag{11}$$

where V_p and V_g are the volumes of polymer matrix and micro-spheres respectively.

According to the series theorem of heat resistance, the effective thermal conductivity of composites, k_{eff} is given by

$$k_{eff} = \left(\frac{H}{RA}\right) = ((H/(R_1 + R_2)A) \tag{12}$$

where, R_1 and R_2 are the heat resistances of Part I and Part II and R is the total resistance of the element.

For micro-spheres we have,

$$V_p\rho_p + V_g\rho_g = V_c\rho_c \tag{13}$$

where V_p and V_g are the volumes of polymer matrix phase and micro-sphere phase respectively and V_c is the Volume of the composite and ϱ_p and ϱ_g are the densities of the polymer matrix phase and micro-sphere phase repectively and ϱ_c is the density of the composite as a whole.

The thermal resistance, $R = d/kA$ (14)

$$R_1 = R_3 = h/k_pA \tag{15}$$

$$R_2 = \frac{2r}{\left(\frac{k_p\ V_p}{2Ar} + \frac{k_g\ V_g}{2Ar}\right)A} = 4r^2/(k_pV_p + k_gV_g) \tag{16}$$

The volume fraction is given by

$$v_f \ = 4\pi r^3/3H^3 \tag{17}$$

where 2r is the particle diameter and H is the length of the side of the square.

Again h_1 = H-2r and h_2 = 2r,

Substituting value of 'H' in the form of volume fraction and h_1 and h_2 in equation (5) we get

$$k_{eff} = \frac{r\left(\frac{4\pi r^3}{3\pi_f}\right)^{\frac{1}{3}}}{\left(\frac{h_1}{k_pA}\right) + \left(\frac{4r^2}{k_pV_p + k_gV_g}\right)} = \frac{r\left(\frac{4\pi r^3}{3\pi_f}\right)^{1/3}}{\left(\frac{H-2r}{k_pA}\right) + \left(\frac{4r^2}{k_pV_p + k_gV_g}\right)} \tag{18}$$

Finally, the expression for effective thermal conductivity of the composite is deduced as:

$$k_{eff} = [1/k_p(1 - \frac{6v_f}{\pi})^{1/3} + 2(k_p(\frac{4\pi}{3v_f})^{1/3} + \pi\left(\frac{2v_f}{9\pi}\right)^{1/3} \times (k_g\ \frac{\rho_s}{\rho_g} - k_p))^{-1}]^{-1} \tag{19}$$

Here, k_p and k_g are the respective heat conductivities of the polymer and the micro-sphere phase, ρ_p and ρ_g are the effective densities of the polymer and the micro-sphere phase respectively, and v_f is the volume fraction of the filler i.e. the SGM in the composite.

Experimental Details

Composite Fabrication

SGMs manufactured from high grade Soda Lime Silica Glass containing SiO_2 around 70%, are free from free Lead, Iron and Silicaand produced by firing crushed glass with subsequent collection and cooling of spheroidal product or by melting the formulated glass batch and subsequent breakup of the free falling molten stream to form small droplets. The spheres are mostly based on the A-Glass composition, although E-Glass spheres are also available. A-Glass is recommended for all polymers except for alkali sensitive resins such as polycarbonate, acetal and PTFE for which E- Glass is recommended. Commonly used mean particle sizes may vary from 200 to 35 μm. The chemical properties of SGM are basically those of their precursor silicate glasses.

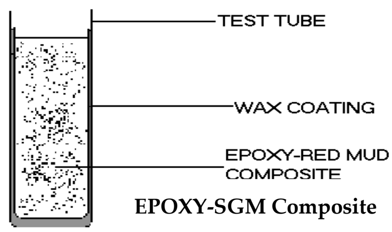
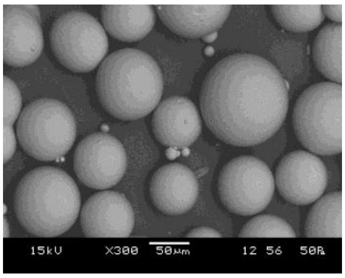


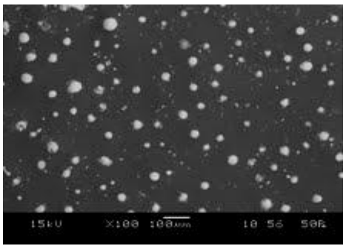
FIG.3 PROCESSING OF SGM-EPOXY COMPOSITE

Spherical glass microspheres of 100, 200 and 300 micron size supplied by Glass Bead Industries India Ltd. are reinforced in epoxy resin to prepare the

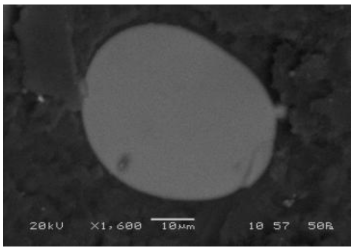
composites. This low temperature curing LY 556 epoxy resin and the corresponding hardener (HY951) are mixed in a ratio of 10:1 by weight as recommended. The dough (epoxy filled with SGM) is then slowly decanted into the glass molds, coated beforehand with wax. The composites are cast in these molds so as to get disc type cylindrical specimens (dia 25 mm, thickness 5 mm). Composites of different compositions (0, 1.4, 3.4, 6.5, 11.3 and 17.9 vol % of SGM for 100 micron size and (0, 6.72, 11.3 and 26.8 vol % of SGM for 200 micron size respectively) are made. The castings are left to cure at room temperature for 24 hours after which the molds are broken and samples are released.



(a) solid glass microspheres used as the filler material



(b) surface morphology of the epoxy-SGM composite



(c) view of a single glass microsphere embedded in the composite

FIG.4 TYPICAL SEM IMAGES OF SGM AND SGM FILLED COMPOSITES

Unitherm™ Model 2022 is used to measure thermal conductivity of a variety of materials. The tests are conducted in accordance with ASTM E-1530 standard. Using the finite-element program ANSYS, thermal analysis is carried out for the conductive heat transfer through the composite body. In order to make this analysis, three-dimensional physical models with spheres-in-cube lattice arrays have been used to simulate the microstructure of composite materials for different filler concentrations.

Numerical Methodology

Using the finite-element program ANSYS, thermal analysis is carried out for the conductive heat transfer through the composite body. In order to make this analysis, three-dimensional physical models with spheres-in-cube lattice arrays have been used to simulate the microstructure of composite materials for five different filler concentrations. Furthermore, the effective thermal conductivities of these epoxy composites filled with SGM up to about 17.9 % by volume for 100 micron SGM are numerically determined using ANSYS.

Results and Discussion

Fig.5. clearly illustrates the heat flow direction and the boundary conditions for the particulate-polymer composite body considered for the analysis of this conduction problem. The temperature at the nodes along the surface ABCD is prescribed as 100^0C and the ambient convective heat transfer coefficient is assumed to be 25 W/m^2-K at a room temperature of 27°C. The other surfaces parallel to the heat flow direction are all assumed adiabatic. The unknown temperatures at the interior nodes and on the other boundaries are obtained with the help of ANSYS.

In this analysis, it is assumed that the composites are macroscopically homogeneous, locally both the matrix and filler are homogeneous and isotropic, in addition, the thermal contact resistance between the filler and the matrix is negligible and the composite lamina is free from voids. The problem is based on spheres-in-cube 3D physical model. The solid glass microspheres are assumed to be uniformly distributed in the matrix, keff of these SGM-epoxy composites is then numerically estimated.

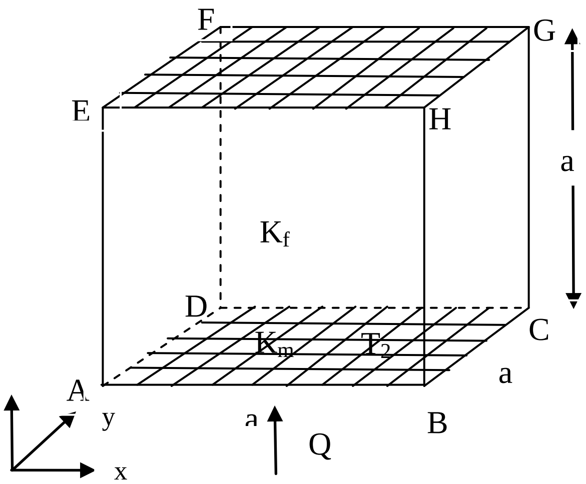


FIG .5 THE HEAT FLOW DIRECTION AND BOUNDARY CONDITIONS

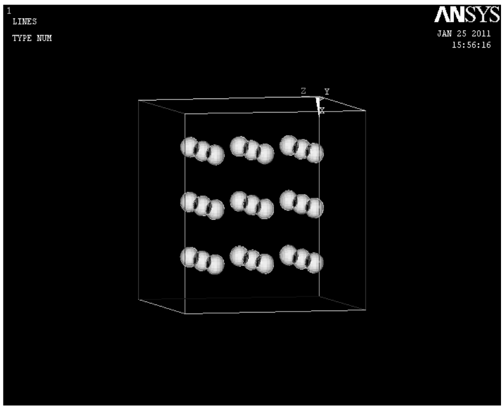


FIG. 6 A TYPICAL 3-D SPHERES-IN-CUBE MODEL WITH FILLER CONCENTRATION OF 1.4 VOL %

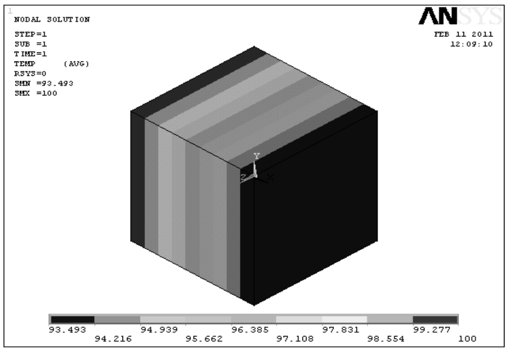


FIG .7 A.TEMPERATURE PROFILE FOR EPOXY-SGM COMPOSITE WITH FILLER CONCENTRATION OF 1.4 VOL %

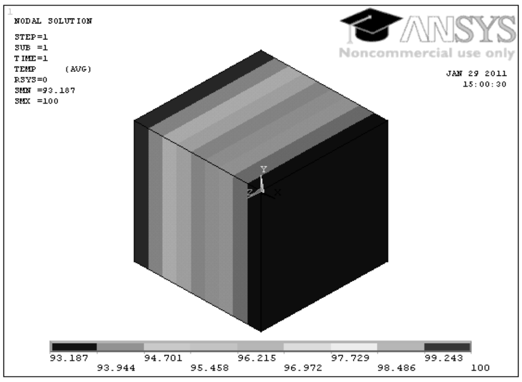


FIG . 7 B. TEMPERATURE PROFILE FOR EPOXY-SGM COMPOSITEWITH FILLER CONCENTRATION OF 3.4 VOL %

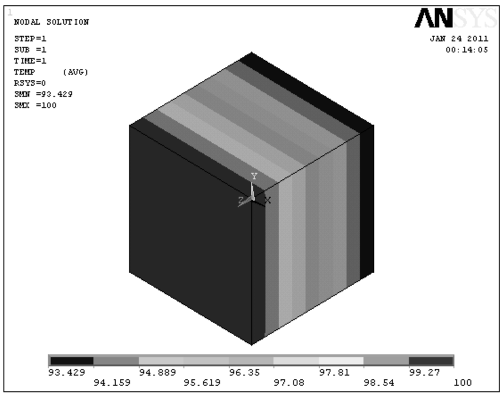


FIG .7 C. TEMPERATURE PROFILE FOR EPOXY-SGM COMPOSITE WITH FILLER CONCENTRATION OF 6.5 VOL %

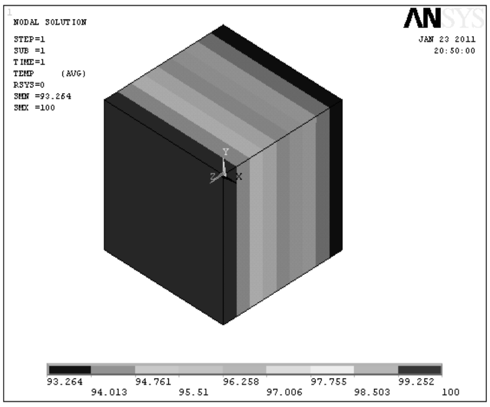


FIG.7D TEMPERATURE PROFILE FOR EPOXY-SGM COMPOSITE WITH FILLER CONCENTRATION OF 11.3 VOL %

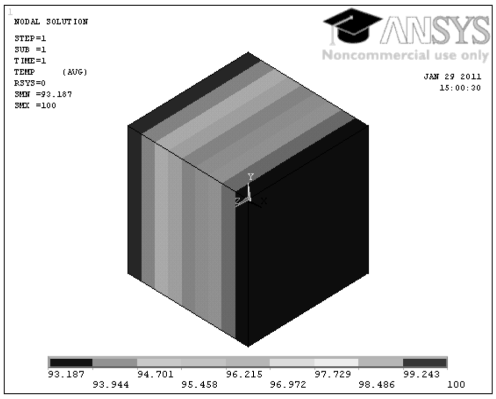


FIG .7 E. TEMPERATURE PROFILE FOR EPOXY-SGM COMPOSITE WITH FILLER CONCENTRATION OF 17.9 VOL %

It is noticed that the results obtained from the finite-element analysis are reasonably closer to the measured values; rather it underestimates the value of effective thermal conductivity for composites of different filler content. However, it leads to a conclusion that for a particulate filled composite of this kind the finite element analysis can very well be used as a predictive tool to determine the effective thermal conductivity of a wide range of particle concentration.

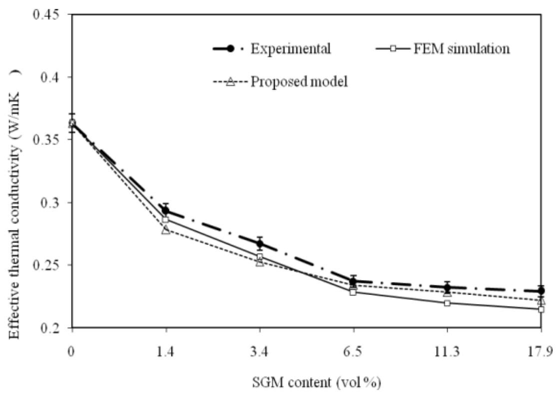


FIG 8 VARIATION OF EFFECTIVE THERMAL CONDUCTIVITY WITH SGM CONTENT: COMPARISON OF THEORETICAL, NUMERICAL SIMULATION AND MEASURED VALUES

Using the theoretical model proposed in this work, the effective thermal conductivity keff of the composites is calculated. The variation of keff with the SGM (100 micron) content is shown in Fig.6 which presents a comparison of the simulated values with the proposed theoretical model as well as experimentally measured ones. It is seen that the results obtained from the proposed correlation are in good agreement with FEM as well as experimental results. The reduction in heat conduction capability is maximum for the SGMs with 100 micron size. It is encouraging to note that the incorporation of SGM results in significant drop in thermal conductivity of epoxy resin. The difference between the simulated values and the measured values of thermal conductivity may be attributed to the fact that some of the assumptions taken for the numerical analysis are not real. However, it is encouraging to note that the incorporation of SGM results in significant drop in thermal conductivity of epoxy resin. With addition of 1.4 vol. % of SGM (100 micron), the thermal conductivity of epoxy 19.283% and with addition of 17.9 vol. % of SGM the conductivity decreases by about 36.9%. Similarly, with the addition of 11.3 vol % of SGM (200 micron), the thermal conductivity drops by 21.2% while for SGM content of 26.8 vol %, it decreases by 30.85%.

Conclusions

Successful fabrication of epoxy based composites filled with solid glass micro-spheres by hand-lay-up technique is possible. A new theoretical model of heat conduction through such composites is developed and a correlation for effective thermal conductivity as afunction of filler content is proposed. The results obtained from this theoretical model are in close approximation to the measured as well as simulated values of effective thermal conductivities for composites with different volume fractions of filler. This work shows that incorporation of SGMs results in reduction of thermal conductivity of epoxy resin and thereby improving its thermal insulation capability. With addition of 17.9 vol % of SGM, the thermal conductivity of neat epoxy decreases by about 36.9%. For the same volume fraction of SGM in the composite, the improvement in composite insulation capability is found to be more for micro-spheres of smaller size range. With light weight and improved insulation capability, SGM filled epoxy composite can be used for applications such as electronic packages, insulation board, food container, thermo flasks, building materials, space flight and aviation industry etc.

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